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A NOTE ON THE USE OF A PHOTOELECTRIC COUNTER FOR TOTAL PARTICLE COUNTING

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Abstract—The ability of the Nolan–Pollak counter to function as a "total" particle counter was investigated. Comparison with the UMR Absolute Aitken Nuclei Counter operated at an expansion pressure ratio of 1.33 revealed that in mean, the N–P counter measurements (at an expansion pressure ratio of 1.21) were 30% less. This correction, however, depends on particle nature, concentration and size distribution and assumes that particle losses in the central part of the UMR-AAN counter are negligible.

INTRODUCTION

Cloud physicists make a distinction between Aitken nuclei (AN) and cloud condensation nuclei (CCN). The first group encompasses all submicron particles $(10^{-7} < r < 10^{-5} \text{ cm})$ activated during an air expansion in a suitable counter just below the critical supersaturation of water vapor corresponding to the activation of small ions. CCN, on the other hand, are activated at supersaturations occurring in nature during cloud and fog formation. The importance of AN counters as "total" particle (nuclei) counters has been stressed often for characterizing the air quality both in heavily polluted and unpolluted areas. The latter use assumes the comparability of counters based on different principles with regard to specific environmental conditions.

Earlier attempts to compare the AN counters are described in the survey article by Pollak (1959), where several important questions, such as counting of all successive showers ("Nachzüglers") in an absolute counter, the efficiency of an AN counter in counting nuclei of different concentration, mean size and specific composition have been discussed. The author summarized attempts by several investigators to estimate different kinds of particle losses in the counters and the dependence of a counter's efficiency on the expansion ratio saying: "For many applications to meteorology a set of nucleus counts at various expansion ratios would be of great interest".

Since the publication of Pollak's survey article in 1959 several attempts to compare AN counters, including three international workshops as well as several laboratory comparisons of both absolute and relative counters have been made. These comparisons were motivated by the necessity to establish a correlation between the very handy relative counters operating by various principles and an absolute counter (Grant, 1971; Saxena *et al.*, 1972; Wegrzyn and Podzimek, 1975; Podzimek and Kassner, 1976; Cadle *et al.*, 1975; Jaenicke and Kanter, 1976; Rosen *et al.*, 1978; Postendörfer and Soderholm, 1978; Plomp *et al.*, 1979; Liu *et al.*, 1979). The counter comparison convincingly displayed the high accuracy and reproducibility of N-P relative counter with 209 cm³ sensitive volume and 2.50 cm i.d. (e.g. Garland and Branson, 1977) which many investigators have started to use as a secondary standard. Furthermore, it became apparent that several counters exhibited discrepancies when test aerosol of a certain composition and size distribution was used (e.g. Wegrzyn and Podzimek, 1975; Cooper and Langer, 1978; Holländer and Schumann, 1979). Several investigations also showed how difficult it is to take a representative sample and stabilize the aerosol without

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losing a considerable part of particles with diameters smaller than 0.004 μ m due to the particle diffusion toward the walls of sampling tubes or fog chambers (e.g. Fürth, 1955; Davies, 1981). One may ask a very legitimate question as to whether these deviations are inherent to a specific counter and test aerosol and what is the mean deviation from a standard counter measurement. There were some speculations regarding the consistently higher counts of the UMR-Absolute Aitken Nuclei Counter (UMR-AANC) when compared to the N–P counter. It was suggested that it can be explained by the higher saturation ratio (3.45) in the UMR-AAN counter (Podzimek and Kassner, 1979) as opposed to 2.29 used in the N–P counter. This question, having—besides particle diffusional losses—a cardinal importance for the use of an AN counter as a "total" ultrafine particle counter, is discussed in this contribution.

RESULTS OF THE AITKEN NUCLEI COUNTER COMPARISON WITH THE UMR-AAN COUNTER

The UMR-AAN counter, with its large sensitive volume (151.) and variable expansion pressure ratio, is a unique instrument for AN counter calibration, and was originally described by Kassner *et al.* (1971). The main specifications of the UMR-AAN counter are mentioned in the appendix. It was used most recently during an AN counter comparison with two N-P counters of the same type, Gardner (GA) counter and General Electric (GE) automatic nuclei counter. The counters were connected with 2.0 m long tygon tube of 1 cm i.d. with an aerosol storage chamber (having inside a mylar bag of 15.0 m³ or 0.5 m³). The volumetric flow rate during flushing and filling N-P counter and UMR-AAN counter was around 10 l./min. For comparison aerosol was vented both straight and through Kr-85 neutralizers into the counters. Particle characteristics were obtained from the electronmicrographs (grids were exposed in the TSI electrostatic sampler) and—e.g. in case of room aerosol—from the TSI screen diffusion battery operated at standard conditions. A detailed description of the experimental setup of the calibration, parameters of the test aerosol and the results of counter comparison are published elsewhere (Podzimek *et al.*, 1979). Here, only several of the most important results will be discussed.

AN counter comparison has been made with sodium chloride, ammonium sulfate, carbon, nichrom wire and room aerosols in the nuclei concentration range between 150 and 8.0×10^4 cm⁻³. However, for better statistical comparability of results obtained from specific aerosols, only concentrations of aerosols up to 2.0×10^4 AN/cm³ were correlated. Regardless of the concentration and median diameter of generated aerosol particles (0.07 μ m for NaCl; 0.065 μ m for (NH₄)₂ SO₄; 0.006–0.15 μ m for carbon; below 0.01 μ m for Ni–Cr wire aerosol and 0.02 μ m for room aerosol) the UMR-AAN counter yielded higher counts than any other. The recorded concentrations exceeded those of the GE counter by 33.6% for NaCl, 29.9% for (NH₄)₂ SO₄, 47.4% for Ni–Cr, 24.5% for carbon and 41.6% for room air. Smaller deviations were found for GA counter: 7.1% for NaCl, 29.9% for (NH₄)₂ SO₄, 32.6% for Ni–Cr; 15.0% for carbon and 20.6% for room air. Large scatter of data was usually found at concentrations below 500 AN/cm³ and above 15,000 AN/cm³ (Podzimek *et al.*, 1981).

Results of a comparison between UMR-AAN counter and the N-P counter 922 for different test aerosols are plotted in Fig. 1. In the same figure a data correlation was made to N-P counter 920 which was originally calibrated against Dr. Pollak's counter in Dublin by Mr. P. Allee from NOAA laboratories at Boulder, Colorado. The "smooth" curves for a specific aerosol represent the regression lines for parabolic curve fitting of the data obtained for different AN concentrations relative to the light extinction measured in the N-P counter. As explained in the appendix, AN absolute concentrations were counted from the photographic record of droplets taken in the UMR-AAN counter. The maximal total error of this evaluation is considered to be lower than 15% of the measured nuclei concentration (Podzimek *et al.*, 1979). After evaluating all measurements, one concludes that the UMR-AAN counter records higher AN concentrations than N-P counter 920 with mean differences



Fig. 1. Comparison between UMR-AAN counter and N-P counter No. 922 for different aerosols and AN concentrations. A dashed curve corresponds to the N-P counter No. 920 data for aged room aerosol.

as high as 30-40% in the AN concentration range corresponding to moderately polluted areas. This indicates that the higher AN counts for the UMR-AAN counter could possibly be explained by the fact that it is operated at a higher saturation ratio than the N-P counter.

NUCLEI CONCENTRATION AT DIFFERENT EXPANSION PRESSURE RATIO

The use of variable expansion pressure ratio in the Large Scholz Counter enabled Foitzig to investigate the increase in AN counts with increasing pressure ratio (Pollak, 1959). However, the investigator did not continue in his study beyond the ratio of 1.20 into the region where concentration of small ions starts to play an important role. In the sphere of relative AN counters the study by Pollak and Metnieks (1959) helped to establish the expansion ratio of 1.21 as a standard operating ratio for N–P counter. Some doubt about the universal validity and suitability of this limit was expressed by Wegrzyn and Podzimek (1975) and by Sinclair (1975). Sinclair thought that expansion ratios as high as 1.26 would better reflect the "total" particle counts and that Pollak and Metnieks (1959) did not use particles small enough to check the counter's performance in counting all ultrafine particles potentially occurring in the atmosphere.

During the last few years the real supersaturation produced in an AN counter was questioned and several times discussed (e.g. by Jaenicke and Kanter, 1976). Pollak and Metnieks (1960) assumed that a pressure ratio of 1.21 corresponds to a supersaturation of 345 %. However, based on the air pressure of 760 mm Hg and a temperature of 20 °C, the pressure ratio of 1.21 is equivalent to a supersaturation ratio of approximately 2.29 (or to a supersaturation of 129 %). Because most of the operators of AN counters are using the calibration against N-P counters as a basis for field measurements, one has to first ask the question: "Is a supersaturation of 129 % in the counter's sensitive volume high enough to count the majority of ultrafine particulates?".

The experimental setup of the investigation of the dependence of AN counts on the N-P counter expansion pressure ratio was similar to the AN studies made earlier in the UMR-laboratory (Podzimek *et al.*, 1981) with the exceptions that only two counters were involved (N-P and GE counter) and a smaller mylar storage bag was used. The GE counter continuously monitored the concentration and stability of the test aerosol in the storage bag. Three different test aerosols were used: Room, NaCl and carbon. The characteristics of room

aerosol (e.g. $d_m \cong 0.02 \ \mu m$) were mentioned earlier. Because of the intended use of low as well as very high AN concentrations, a special generating and aerosol sampling technique was used for NaCl and carbon aerosol.

NaCl aerosol was generated in a nitrogen atmosphere by the evaporation-condensation technique using two electrically heated ovens: The first, in which a ceramic boat with reagent grade NaCl crystals was placed, was at 715 °C and the second at 605 °C. The ovens were about 13 cm apart connected by a 2.54 cm i.d. ceramic pipe which was not thermally insulated. Temperatures were measured by thermocouples placed at the outer wall of the ceramic tube passing the vapors and aerosol through the ovens. The setup (utilizing a particle charge neutralizer) enabled extremely high particle concentration output of more than 10⁶ AN/cm³ at a flow rate of 7 l./min. The particle median diameters were around 0.06 μ m and the dispersity factors $\alpha_g = \sigma_g/r_g = 0.59$. Different aerosol concentrations were obtained by means of a diluter (Environmental, SN 7401) which diverts part of the aerosol stream through a series of absolute filters. This device contributed also to the damping of the intense concentration fluctuations of the freshly generated aerosol.

The carbon aerosol was generated by evaporating electrically heated carbon rods in an apparatus similar to that used for coating specimens for electronmicroscopy. In the main container, however, a continuous nitrogen gas flow of 15 l./min was maintained instead of a vacuum. Use of the aerosol neutralizer and diluter resulted in aerosol concentrations between 1.0×10^3 and 5.0×10^4 AN/cm³. The particle median diameter of the aged carbon aerosol was around 0.05 μ m with the dispersity factor $\alpha_q = \sigma_q/r_q = 0.32$.

The following procedure for operation of the N–P counter was observed (Metnieks and Pollak, 1959): The N–P counter was flushed for at least 1 min at a flow rate of 71./min with aerosol laden air passing through 150 cm long tygon tube of 1.0 cm i.d. before the sample was taken. The airflow entered the counter from the top and was drawn out from the bottom. After the sample was introduced to the counter filtered nitrogen gas was used for raising the internal pressure of 160 mm Hg. A 1 min waiting period was found sufficient to dissipate the heat due to the gas compression and humidify the gas. The GE counter reading was taken when the expansion of the air in the N–P counter was performed and the needle of the microammeter reached its minimum value.

The mentioned procedures were applied to an interesting experiment with room aerosol in the concentration range of 3000–25,000 AN/cm³ which was performed with the intent of concentration curve comparison with the curve for spontaneous nucleation in the counter. The curves (Fig. 2) for room aerosol were obtained by sampling the laboratory air in the N–P counter and successively increasing the sample pressure with filtered nitrogen gas. The curve for spontaneous nucleation (SN) in the same figure is based on nuclei counting (extinction measurement) in filtered nitrogen at high pressure expansion ratio. The large scatter of points in the spontaneous nucleation curve is probably due to the unfavorable shape of the N–P counter's sensitive volume (large surface volume ratio) influencing the heat transfer from the wall, diffusion and the homogeneity of the nuclei (especially around the valve inlets at high expansion pressure ratio).

For filtered nitrogen the sharp increase in nuclei counts started at the pressure expansion ratio of 1.36, and at 1.66 the AN concentration surpassed 20,000 AN/cm³. Similar findings were made with filtered air, where a markable increase in AN counts started at an expansion ratio of 1.32. During this experiment the "background" aerosol measured by the GE counter constantly showed counts far below 100 AN/cm³. Room aerosol was characterized by a "plateau" above the recommended expansion pressure value for the N-P counter of 1.21. This difference can amount to 10% in case of nuclei concentrations around 6000 AN/cm³, and up to 20% in case of concentrations around 13,000 AN/cm³. There is a clear tendency toward large increases in nuclei counts if the expansion pressure ratio is approaching that of spontaneous nucleation. However, a higher expansion ratio than that for pure spontaneous nucleation is always required for starting the spontaneous nucleation in presence of room aerosol. One can speculate about the competition among differently sized embryos for a limited amount of water vapor available during air expansion. This might lead to the conclusion that for higher concentrations of AN, higher expansion pressure ratios are



Fig. 2. AN concentration (room aerosol) in dependence of different expansion pressure ratios (E.P.R.) in the N-P counter. SN-curve for spontaneous nucleation in filtered nitrogen gas. Dashed horizontal lines denote the background AN concentration as measured by the calibrated GE counter.



Fig. 3. NaCl aerosol concentration in dependence of different E.P.R. in the N-P counter. Dashed horizontal lines denote the AN background concentration.



Fig. 4. NaCl aerosol concentration in dependence of different E.P.R. in the N-P counter for mean AN concentrations around 30,000 and 65,000 cm⁻³ (horizontal dashed lines).

required to initiate spontaneous nucleation. This simplistic picture must be, however, modified in regard to the effect of differently sized embryonal droplets on light attenuation in the N-P counter.

Measurements with NaCl aerosol of concentration range 2×10^3 to 7.0×10^4 AN/cm³ had results similar to that for room aerosol. At low AN concentrations (2400 AN/cm³) the curve



Fig. 5. Carbon aerosol concentration in dependence of different E.P.R. in the N-P counter for mean AN concentrations around 5500 and 12,000 cm⁻³ (horizontal dashed lines).

has a plateau between expansion pressure ratios 1.27 and 1.50 and a transition into spontaneous nucleation around the ratio of 1.6. The plateau is approximately 20% above the AN concentration measured at an expansion pressure ratio of 1.21. The AN concentration difference between the "plateau" and that corresponding to the ratio of 1.21 increases with the mean AN counts (e.g. for 69,180 AN/cm³ it means an increase of almost 60% of the value at 1.21). Simultaneously, the transition into the regime of spontaneous nucleation is delayed beyond the expansion pressure ratio of 1.65 (Figs 3 and 4).

Carbon aerosol showed a transition into spontaneous nucleation at the ratio of 1.575 if the AN concentration was around 2000 AN/cm³ (Figs 5 and 6). At higher concentrations the transition was shifted beyond the ratio of 1.6. The AN concentration difference between the "plateau" and that corresponding to the ratio of 1.21 is small in case of AN concentrations around 2000 AN/cm³ (10%) and increases to several tens of percent if the concentration is close to 50,000 AN/cm³.



Fig. 6. Carbon aerosol concentration in dependence of different E.P.R. in the N-P counter for mean AN concentrations around 2000 and 55,000 cm⁻³ (dashed lines).

CONCLUSIONS

The systematic investigations of the dependence of AN concentrations on the expansion pressure ratio of the N-P counter lead to the conclusion that the recommended 1.21 ratio is not very appropriate and can lead to the underestimation of the concentration of AN. This difference can amount for nuclei larger than 0.2 μ m in median diameter to 10-20% for very clean air, 30-40% for moderately polluted air, and up to 50-100% in a highly polluted atmosphere. In regard to the use of a N-P counter as a "total" particle counter, it is suggested to increase the standard expansion pressure ratio to 1.30 and/or to use an appropriate correction. Measurements supporting this point are the results described by Wegrzyn and Podzimek (1975) and by Podzimek and Kassner (1979). In case of the use of particles with diameters smaller than 0.01 μ m N-P counter—as well as the other AN counters—will count differently than what has been stressed by several authors earlier. The response of different

counters will depend on the concentration, size distribution and physico-chemical properties of the nuclei, air expansion rate and expansion speed, geometry of the fog chamber and efficiency of the humidification system. Theoretical calculations assume a homogeneously dispersed aerosol and thermal equilibrium. An example, illustrating the applicability of the calculated diffusional losses in N-P counter (Davies, 1981) for the measured room aerosol losses during successively increasing waiting times before air expansion (Podzimek *et al.*, 1979) is presented in Fig. 7. The room aerosol was characterized by having 50% of particulates with $d < 0.015 \,\mu$ m and 10% with $d < 0.005 \,\mu$ m as measured by the screen diffusion battery. Comparison of the experimental data with the two curves calculated by Davies (1981) for particles with radii $r = 0.0015 \,\mu$ m and $r = 0.003 \,\mu$ m shows the usefulness of the model calculation for estimating the ultrafine particle losses which override those stemming from insufficient expansion ratio. For comparison two particle loss curves for Scholz (S) and Kanter (K) counters from the article by Jaenicke and Kanter (1976) are plotted in the same diagram.



Fig. 7. Particle diffusion losses in AN counters: Curves 1 and 2—calculated losses in a N-P counter for particles with radii r = 0.0015 and 0.003 μ m. Concentrations were calculated (Davies, 1981) for the distance x/R = 0.6 from the central line of the fog chamber which is a cylinder with a radius R= 1.27 cm i.d. Curve 3—our experimental data for room air aerosol losses in a N-P counter. S and K—curves corresponding to room aerosol losses in a Scholz and Kanter counter as measured by Jaenicke and Kanter (1976).

A question which remains still unanswered is the nature of the particulates or molecular clusters effective at expansion ratios close to the nucleation of ions in the AN counter. Based on the experiments, one is inclined to believe that their concentration does not decrease so fast as would be predicted by a model of diffusional particle losses or recombination of ions. We would like to investigate further this interesting problem in the future.

This short note presents a very condensed summary of a direct comparison between the UMR-AAN counter and several other types of AN counters (Podzimek *et al.*, 1981). No emphasis was put on the use of monodisperse aerosol because of the potential application of AN counters in the field. The authors of this contribution are aware of the almost impossible task of universal calibration of AN counters. They observed systematic differences in AN counts when aerosol particles of the same mean size, but different nature and concentration,

were used (Podzimek *et al.*, 1981). In this point, their findings deviate from the general conclusion from the experiments made by Cooper and Langer (1978) where no appreciable difference in AN counting in GE counter was found for particle diameters larger than $0.02 \mu m$. They feel the current theory explaining the activation of nuclei of different physico-chemical nature to be inadequate when applied to a fast expansion in a specific sensitive volume. These difficulties are magnified in the relative counters through lack of a reliable model for light extinction in a specific counter.

This rather pessimistic conclusion is not meant to demean the usefulness of AN counters applied in the field for the characterization of the mean level of concentration of submicron particles—if one can tolerate potential concentration deviations of 50 % from the "true value" in moderately polluted air and if it is not essential to know about particulates smaller than 0.005 μ m in diameter.

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APPENDIX. SPECIFICATIONS OF THE UMR-AANC

Operating principle: Air sample (saturated) adiabatic expansion in a Wilson type chamber with distilled water above the mobile bottom (piston with a rubber diaphragm).

Chamber dimensions: Cylinder with i.d. \cong 34.0 cm and $h \cong$ 16.5 cm and total volume $V \cong$ 15,000 cm³.

Supersaturation range: Variable (subject to easy adjustment) between 24 and 280 %.

Counting technique: Direct photography (from the top of the chamber) of droplets in the sensitive volume illuminated horizontally in a beam of 1.5 cm width by a cylindrical flash tube with collimator. Motor-driven

Nikon camera with Micro-Nikkor auto 1:3.5; f = 55 mm lens with shutter speed 1/30 to 1/60 sec. Counting of droplets in photographs projected on a screen using a mechanical counter.

AN concentration range: Dilution of air sample enables to count AN (droplet) concentrations from 10 cm⁻³ to 1,000,000 cm⁻³.

Accuracy of AN concentration measurement: $\pm 10\%$ for AN concentrations around 100 cm^{-3} ; $\pm 15\%$ for 5000 cm⁻³ and $\pm 20\%$ for concentrations larger than 20,000 cm⁻³.

Frequency of measurement: 5 min.

Data output: (a) Role of the film with droplet pictures, (b) 8-inch wide galvanometer-oscillograph record of the pressure before and after expansion, volume of the expansion and of the sample vented in, (c) adjusted temperature of the chamber in terms of the divisions of the meters.

Data processing: WANG electronic calculator or other computer was used for calculating the supersaturation and other thermodynamic parameters.

Number of units: Main chamber: $76 \times 76 \times 109$ cm

Programming unit: $56 \times 48 \times 23$ cm Pressure gauge: $41 \times 38 \times 23$ cm Recorder: $53 \times 41 \times 23$ cm

Power: 115 VAC, 60 cycles, 2.5 kW.